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2.	Patent application number *(The Patent Office will fill in this part)*	**9827699.1** 16 DEC 1998
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	Patents ADP number *(if you know it)*	6166441002
	If the applicant is a corporate body, give the country/state of its incorporation	United Kingdom
4.	Title of the invention	ORGANIC LIGHT-EMITTING DEVICES
5.	Name of your agent *(if you have one)*	
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6.	If you are declaring priority from one or more earlier patent applications, give the country and the date of filing of the or of each of these earlier applications and *(if you know it)* the or each application number	Country / Priority application number *(if you know it)* / Date of filing *(day / month / year)*
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Translations of priority documents	
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Request for preliminary examination and search (Patents Form 9/77)	
Request for substantive examination (Patents Form 10/77)	
Any other documents *(please specify)*	

11. I/We request the grant of a patent on the basis of this application.

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PAGE WHTE & FARRER

Date

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ORGANIC LIGHT-EMITTING DEVICES

This invention relates to organic light-emitting devices (OLEDs) and a method for improving the uniformity of current density of OLEDs having a light-emissive organic layer containing intrinsic defects.

Organic light-emitting devices such as described in US Patent No. 5,247,190 or in US Patent No. 4,539,507, the contents of which are incorporated herein by reference, have great potential for use in various display applications. According to one method, an OLED is fabricated by coating a glass or plastic substrate with a transparent first electrode (anode) such as indium tin oxide (ITO). At least one layer of a thin film of an electroluminescent organic material is then deposited prior to a final layer which is a film of a second electrode (cathode) which is typically a metal or alloy.

In many practical applications, the layer of electroluminescent organic material has a thickness of the order of 100nm in order to ensure a practical operating voltage. It is typically deposited on the first electrode by a spin-coating technique. If the organic material is contaminated with particles having a size of the order of the thickness of the organic layer, not only will these particles themselves give rise to defects in the resulting organic layer, their presence disrupts the movement of the fluid organic material over the surface of the first electrode layer leading to variations in the thickness of the resulting organic layer about the particle, and in the worst case leading to the formation of holes in the organic layer through which the underlying layer (electrode layer) is exposed.

Defects in the organic layer can also be caused by, for example, inherently poor film-forming properties of the organic material, or by physical damage to the organic layer after deposition.

A typical defect site is shown in Figure 3. The electroluminescent organic layer 106 has been deposited by spin coating on a glass substrate 102 coated with an indium tin oxide (ITO) anode layer. The existence of a large particle 107 has led to a defect site 109 comprising the particle 107 itself and a pinhole 111. A cathode layer 110 is formed over the electroluminescent organic layer 106.

Localised defects of the kind shown in Figure 3 can manifest themselves during device operation as a current anomaly (short) where a large proportion of the current becomes localised in the area of the defect. This leads, inter alia, to problems of device reproducibility and is a particular problem in dot matrix devices since it provides alternative current paths that lead to the wrong pixels being lit.

In order to prevent these kind of defects, the deposition of the organic layer is typically carried out in a clean room with a view to preventing contamination and typically involves filtering the organic material prior to spinning to remove large particles therefrom. However, a typical clean room has particle size levels specified down to 300nm and the organic material is only typically filtered to about 450nm, since the elimination of particles having smaller sizes requires great expense.

The light-emissive organic material will therefore often still be contaminated with particles having a size of the order of the thickness of the organic layer to be deposited, which will, as mentioned above, lead to defects in the resulting organic layer. Furthermore, even if the contamination by such large particles could be completely eliminated, defects can still arise during the manufacturing process as a result, for example, of inherently poor film-forming properties of the organic material itself, or due to physical damage inadvertently inflicted on the organic layer after deposition.

One known technique of removing the defect particles after production of the device is by passing a very high current through the device to "burn-out" the defect particles by vaporizing them. However, this technique is not applicable to all defect particles and cannot be used to resolve the problem of large shorts. Moreover, it does not necessarily deal with problems that may manifest themselves in the lifetime of the device.

It is therefore an aim of the present invention to reduce the problem of current anomalies in an organic light-emitting device.

According to a first aspect of the present invention there is provided an organic light-emitting device comprising a light-emissive organic layer interposed between first and second electrodes for injecting charge carriers into the light-emissive organic layer, at least one of said first and second electrodes comprising a plurality of layers including a first electrode layer having a high resistance adjacent the surface of the light-emissive organic layer remote from the other of the first and

second electrodes, said first electrode layer comprising a high-resistance material selected from the group consisting of a mixture of a semiconductor material with an insulator material, a mixture of a semiconductor material with a conductor material and a mixture of an insulator material with a conductor material.

The use of a layer of the above-mentioned mixtures of materials as the high resistance electrode layer has the advantage that the resistance of the high resistance electrode layer can be easily adjusted to the desired value by simply adjusting the relative proportions of the components of the mixture accordingly.

In the case of a cathode, the first electrode layer preferably comprises at least one material having a low work function, preferably less than 3.7 eV, and further preferably less than 3.2 eV, to improve the electron-injecting performance of the cathode.

According to a second aspect of the present invention, there is provided an organic light-emitting device comprising a light-emissive organic layer interposed between first and second electrodes for injecting charge carriers into the light-emissive organic layer, at least one of said first and second electrodes being opaque and comprising a plurality of layers including a thin first electrode layer comprising a low work function material adjacent the surface of the light-emissive organic layer remote from the other of the first and second electrodes, and a second electrode layer adjacent the surface of the first electrode layer remote from the organic light-emissive material, said second electrode layer comprising a layer of a high-resistance material comprising a semiconductor material, a mixture of a

semiconductor material with an insulator material, a mixture of a semiconductor material with a conductor material or a mixture of an insulator material with a conductor material.

The thin first electrode layer in this second aspect of the present invention preferably has a thickness in the range of 0.5 to 10 nm, and is further preferably 5nm or less.

According to a third aspect of the present invention, there is provided an organic light-emitting device comprising a light-emissive organic layer interposed between first and second electrodes for injecting charge carriers into the light-emissive organic layer, at least one of said first and second electrodes comprising a plurality of layers including a first electrode layer having a high resistance, said first electrode layer having a thickness greater than the light-emissive organic layer, such that any intrinsic defects in the light-emissive organic layer are covered by the first electrode layer.

According to one embodiment, the first electrode layer is disposed adjacent the surface of the light-emissive organic layer remote from the other of the first and second electrodes.

By making the thickness of the high resistance first electrode layer greater than that of the light-emissive organic layer, any pinhole defects in the light-emissive organic layer are completely filled making it possible to further ensure that there

are no areas of the light-emissive organic layer left exposed to make direct contact with an overlying conductive layer.

The high resistance layer in this third aspect of the invention preferably comprises a semiconductor material, a mixture of a semiconductor material with a conductor material, a mixture of a semiconductor material with an insulator material or a mixture of a conductor material with an insulator material.

According to yet another aspect of the present invention, there is provided a method for improving the uniformity of current density of an organic light-emitting device comprising a light-emissive organic layer interposed between first and second electrodes for injecting charge carriers into the light-emissive organic layer, the method comprising the step of forming one of the first and second electrodes from a plurality of layers including a first electrode layer having a high resistance comprising a semiconductor material, a mixture of an insulator material with a semiconductor material, a mixture of an insulator material with a conductor material, or a mixture of a semiconductor material with a conductor material.

In each of the above aspects of the invention, the high-resistance electrode layer is preferably capped with a layer of a conductor material such as a layer of aluminium.

The resistance of the high resistance electrode layer is preferably selected such that it is not too high to cause a significant increase in the drive voltage (since this will reduce the power efficiency of the device) but is high enough to prevent

excessive currents at defect sites. Typically for an electrode layer of thickness lying in the range of 100-10000nm, the resistivity lies in the range 1 to 10^5 Ωcm.

Suitable semiconductor materials for use in the present invention include, for example, Ge, Si, α-Sn, Se, ZnSe, ZnS, GaAs, GaP, CdS, CdSe, MnS, MnSe, PbS, ZnO, SnO, TiO_2, MnO_2 and SiC.

Suitable insulator materials for use in the present invention include, for example, insulating oxides, nitrides and fluorides such as Al_2O_3, SiO_2, LiO, AlN, SiN, LiF and CsF. Suitable conductor materials for use in the present invention include, for example, metals such as Al and Ag.

Suitable low work function materials for use in the present invention include, for example, Ca, Li, Yb, LiF, CsF and LiO.

The use of the cathode to combat the undesirable effects of intrinsic defects in the light-emissive organic layer is particularly advantageous when the cathode is deposited in a vacuum because of the ability to keep particulate levels extremely low.

Embodiments of the present invention will now be described, by way of example only, with reference to the accompanying drawings in which:-

Figure 1 is a cross-sectional view of an OLED according to an embodiment of the present invention.

Figure 2 is a cross-sectional view of an OLED for explaining the principle of the present invention.

Figure 3 is a schematic cross-sectional view of an OLED having a typical defect site caused by particulate contamination of the organic material during spin-coating.

Figure 4 is a cross-sectional view of an OLED according to a second embodiment of the present invention.

Figure 1 shows an OLED according to a first embodiment of the present invention. A glass substrate 2 having a thickness of 1.1mm is coated with a layer 4 of indium tin oxide (ITO) with a sheet resistance of 15 Ohms/sq. to a thickness of 150nm. Although not shown in Figure 1, this is patterned to form a series of parallel strips using, for example, standard photolithographic and etch processes. A layer 6 of polyethylenedioxythiophene doped with polystyrene sulphonic acid (PEDT:PSS) is spun on the anode layer 4 and subsequently baked at 150°C to remove water leaving a layer of 50nm thickness. A layer 8 of a light-emissive polymer such as a blend of 5% poly(2,7-(9,9,di-n-octylfluorene)-3,6-(benzothiadiazole) with 95% poly(2,7-(9,9-di-n-octylfluorene) (5BTF8) doped with poly(2,7-(9,9-di-n-octylfluorene)-(1,4-phenylene-((1,4-phenylene-((4-secbutylphenyl)imino)-1,4-phenylene)) (TFB) is then spun on to the layer 6 of PEDT:PSS to a thickness of 75nm. A cathode layer 10 is then formed on the layer 8 of light-emissive polymer.

A standard vacuum evaporation technique is used to deposit the cathode layer in view of the fact that, being a relatively low-energy technique, it causes minimal damage to the underlying layer of light-emissive polymer. If the possibility of

damage to the underlying organic layer is not a concern, sputtering is a desirable technique because it is a conformal deposition technique. In the case of sputtering, neon is preferably used as the discharge gas.

In this case, the cathode layer 10 is a layer of LiF co-evaporated with Al. This cathode layer 10 is deposited to a thickness of between 0.5 and 1 micron to ensure that the entire surface of the underlying organic layer, and hence any defects therein, is covered by the cathode layer.. A layer 12 of aluminium is deposited on top of this layer to a thickness of 0.5 microns. This top layer of aluminium 12 can, for example, be deposited by evaporation. Although not shown in Figure 1, the cathode comprising the LiF-Al cathode layer 10 and the aluminium top layer 12 is also patterned in the form of a series of parallel strips running in a direction orthogonal to the series of parallel anode strips, whereby an ordered array of pixels is formed defined by the points at which each series of cathode and anode strips overlap.

The LiF has a dual function. It is a low work function material and therefore assists the injection of electrons into the light-emissive organic layer. It is also an insulator resulting in a layer having a high resistance.

The high-resistance LiF/Al layer conducts via a percolation mechanism.

The relative proportions of LiF and Al in the LiF-Al layer 10 will be determined according to the desired resistivity. The desired resistivity is itself determined according to the number and area of the defects existing in the underlying light-

emissive organic layer 8. A method for determining a suitable resistivity is described below with reference to Figure 2 which shows an OLED comprising a light-emissive organic layer 18 containing a plurality of pinhole defects 30 which are the major cause of current anomalies in OLEDs.

The light-emissive organic layer 18 is sandwiched between a first cathode layer 20 and an ITO anode layer 14 coated on a glass substrate 12. The first cathode layer is coated with a layer of aluminium 22.

It is supposed, by way of example, that the current density (j) of the device at a typical operating voltage of 3V would be $1 mA/cm^2$ if the light-emissive organic layer 18 did not contain any pinhole defects.

It is desired that the current density attributable to the existence of the pinhole defects represents only a small proportion of the current density that would be observed if there were no pinholes existing in the light-emissive organic layer. For example, it is preferred that the first cathode layer is of a sufficiently high resistance that the current density attributable to the defects is at most 1% of the current density that would be observed if there were no pinholes existing in the light-emissive organic layer.

The current density through the pinhole defects can be calculated to be:

$j_{(def)} = NVA/\rho t$

where N is the density of defects (per unit area); A is the average area of each defect; V is the operating voltage; ρ is the resistivity of the cathode layer; and t is the thickness of the first cathode layer 20.

Let us now suppose that the thickness of the first cathode layer 20 is 0.5 microns, and that there are 100 defects each of area 1μm^2.

Then, at the operating voltage of 3V mentioned above, the current density attributable to the defects would be approximately 60/ρ mA/cm^2.

In order for this current density to represent 1% or less of the current density that would be observed if there were no pinhole defects (which is supposed as above to be 1mA/cm^2), the resistivity of the material of the first cathode layer would have to be about 6000Ωcm or greater.

The voltage drop across a first cathode layer having a thickness of 0.5 microns and composed of material having a resistivity of 6000Ωcm would only be about 0.3mV when the current density is 1mA/cm^2. This layer will therefore have a negligible effect on the power efficiency whilst improving the uniformity of the current density of the OLED in operation.

The existence of particle defects in the light-emissive organic layer have been ignored on the basis that their effect is negligible compared to that of the pinhole defects. However, if the effects of any such defects are not negligible, it will be clear to the skilled person in light of the above how to take the effect of such

particle defects into consideration when determining a suitable resistivity for the high resistance cathode layer.

Figure 4 is a cross-sectional view of an organic light-emitting device according to a second embodiment of the present invention. The substrate 202, anode layer 204, organic layers 206, 208 are identical to those of the first embodiment described above. A thin layer of calcium 209 having a thickness of 5nm is formed on the surface of the organic layer 208. This layer 209 is preferably formed by vacuum evaporation. A layer of silicon 210 having a thickness of 0.5 microns is formed on the thin layer of calcium 209 as a high-resistance layer, and a layer of aluminium 212 having a thickness of 0.5 microns is formed on top of the layer of silicon 210.

The use of a thin layer of a conductor material (in this case, calcium) between the high-resistance layer and the light-emissive organic layer is advantageous in one respect in that it can assist the burning-out of defects in the underlying organic layer by passing a high current through the device after production is completed.

Although, the present invention has been described in detail above with reference to the formation of a high-resistance cathode, it is equally applicable to the formation of a high-resistance anode in the case that an OLED is produced by first forming a cathode on a glass substrate, depositing a layer of light-emissive organic material on the cathode by spinning, and finally forming an anode on the light-emissive organic layer. In the case of an anode, it is preferred that the high-resistance electrode layer comprises a high work function material, or that a thin

layer of a high work function material is interposed between the high-resistance electrode layer and the light-emissive organic layer.

CLAIMS

1. An organic light-emitting device comprising a light-emissive organic layer interposed between first and second electrodes for injecting charge carriers into the light-emissive organic layer, at least one of said first and second electrodes comprising a plurality of layers including a first electrode layer having a high resistance adjacent the surface of the light-emissive organic layer remote from the other of the first and second electrodes, said first electrode layer comprising a high-resistance material selected from the group consisting of a mixture of a semiconductor material with an insulator material, a mixture of a semiconductor material with a conductor material and a mixture of an insulator material with a conductor material.
2. An organic light-emitting device according to claim 1 wherein the first electrode layer comprises at least one material having a low work function.
3. An organic light-emitting device according to any preceding claim wherein the semiconductor material is selected from the group consisting of Ge, Si, α-Sn, Se, ZnSe, ZnS, GaAs, GaP, CdS, CdSe, MnS, MnSe, PbS, ZnO, SnO, TiO_2, TiO_2, MnO_2 and SiC.
4. An organic light-emitting device according to any preceding claim wherein the insulator material is selected from the group consisting of an oxide, a nitride and a fluoride.
5. An organic light-emitting device according to any preceding claim wherein the insulator material is selected from the group consisting of Al_2O_3, SiO_2, LiO, AlN, SiN, LiF and CsF.

6. An organic light-emitting device according to any preceding claim wherein the conductor material is a ductile metal.

7. An organic light-emitting device according to any preceding claim wherein the conductor material is selected from the group consisting of Al and Ag.

8. An organic light-emitting device according to any preceding claim wherein the first electrode layer is comprised of a mixture selected from the group consisting of LiF/Al, Ca/Ge, Li/Si, Ca/ZnO, LiF/ZnSe and CsF/ZnS.

9. An organic light-emitting device according to any preceding claim wherein the organic light-emitting device further comprises a second electrode layer on the first electrode layer, said second electrode layer comprising a layer of a conductor material.

10. An organic light-emitting device according to claim 9 wherein the second electrode layer comprises a layer of a ductile metal.

11. An organic light-emitting device according to claim 1 wherein the thickness of the first electrode layer is in the range of 0.5 to 1.0 microns.

12. An organic light-emitting device comprising a light-emissive organic layer interposed between first and second electrodes for injecting charge carriers into the light-emissive organic layer, at least one of said first and second electrodes being opaque and comprising a plurality of layers including a thin first electrode layer comprising a low work function material adjacent the surface of the light-emissive organic layer remote from the other of the first and second electrodes, and a second electrode layer adjacent the surface of the first electrode layer remote from the light-emissive organic layer, said second electrode layer comprising a layer of a high-resistance material selected from the group consisting of a semiconductor material, a mixture of a semiconductor

material with an insulator material, a mixture of a semiconductor material with a conductor material and a mixture of an insulator material and a conductor material.

13. An organic light-emitting device comprising a light-emissive organic layer interposed between first and second electrodes for injecting charge carriers into the light-emissive organic layer, at least one of said first and second electrodes comprising a plurality of layers including a thin first electrode layer comprising a high work function material adjacent the surface of the light-emissive organic layer remote from the other of the first and second electrodes, and a second electrode layer adjacent the surface of the first electrode layer remote from the organic light-emissive material, said second electrode layer comprising a layer of a high-resistance material selected from the group consisting of a semiconductor material, a mixture of a semiconductor material with an insulator material, a mixture of a semiconductor material with a conductor material and a mixture of an insulator material with a conductor material.

14. An organic light-emitting device according to claims 12 or 13 further comprising a third electrode layer on the surface of the second electrode layer remote from the first electrode layer, said third electrode layer comprising a conductor material.

15. An organic light-emitting device according to claim 14 wherein the third electrode layer is comprised of a ductile metal.

16. An organic light-emitting device according to claim 12 wherein the first electrode layer is comprised of a layer of a material selected from the group consisting of Ca, Li, Yb, LiF, CsF and LiO.

17. An organic light-emitting device according to claim 12 or claim 13 wherein the thickness of the first electrode layer is in the range of 0.5nm to 10nm, preferably less than 5nm.

18. An organic light-emitting device comprising a light-emissive organic layer interposed between first and second electrodes for injecting charge carriers into the light-emissive organic layer, at least one of said first and second electrodes comprising a plurality of layers including a first electrode layer having a high resistance, said first electrode layer having a thickness greater than the light-emissive organic layer, such that any intrinsic defects in the light-emissive organic layer are covered by the first electrode layer.

19. An organic light-emitting device according to claim 18 further comprising a second electrode layer adjacent the surface of the first electrode layer remote from the light-emissive organic layer, said second electrode layer comprising a layer of a conductor material.

20. An organic light-emitting device according to claims 18 or 19 wherein the thickness of the first electrode layer is in the range of 0.5 to 1 micron.

21. An organic light-emitting device according to claim 18 wherein the first electrode layer comprises a material selected from the group consisting of a semiconductor material, a mixture of a semiconductor material and an insulator, a mixture of a semiconductor material and a conductor material and a mixture of an insulator material and a conductor material.

22. A method for improving the uniformity of current density of an organic light-emitting device comprising a light-emissive organic layer interposed between first and second electrodes for injecting charge carriers into the light-emissive organic layer, the method comprising the step of forming one of the first and

second electrodes from a plurality of electrode layers including a first electrode layer having a high resistance, said first electrode layer comprising a material selected from the group consisting of a semiconductor material, a mixture of a semiconductor material with an insulator, a mixture of a semiconductor material with a conductor material and a mixture of an insulator material with a conductor material.

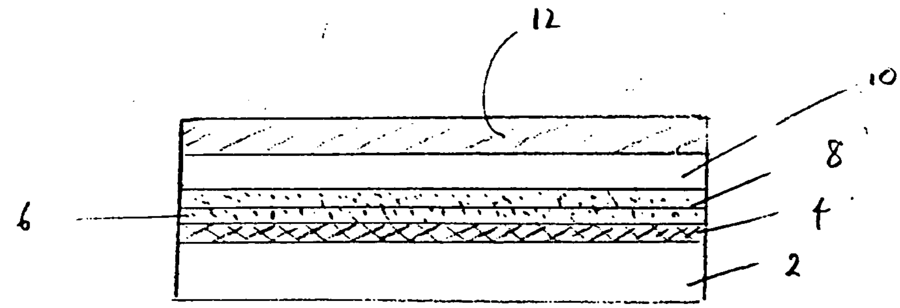

FIG. 1

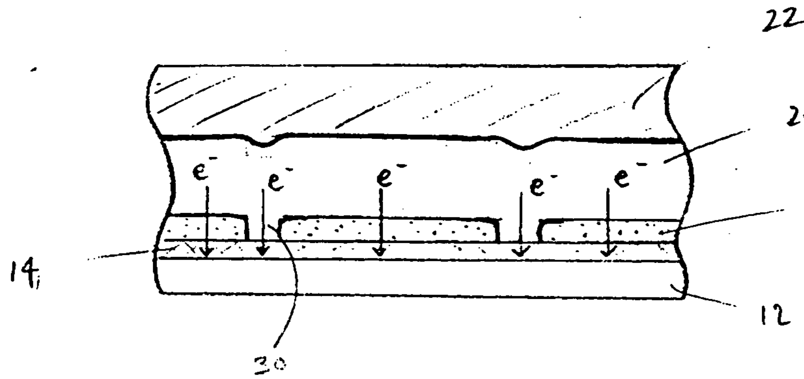

FIG. 2

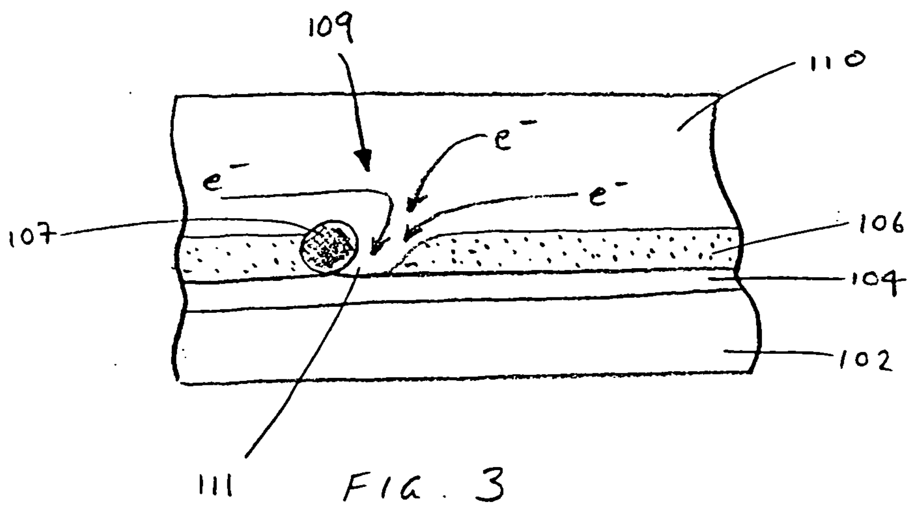

FIG. 3

212.

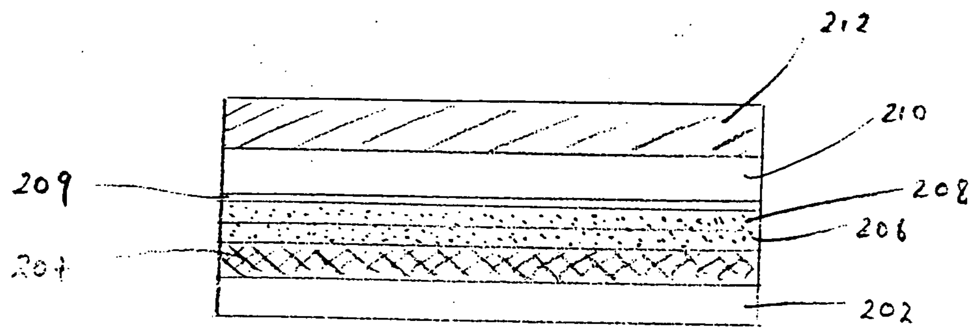

FIG. 4